

PATENT APPLN. NO. 09/524,575  
SUBMISSION UNDER 37 C.F.R. § 1.114

**PATENT**

**REMARKS**

Table 2 has been amended to correct a typographical error. Specifically, in Example 7 "Adsorbent-catalyst E" has been amended to --Adsorbent-catalyst C--. A person of ordinary skill in the art would recognize that "Adsorbent-catalyst E" in Example 7 in Table 2, an example within the scope of the present invention, is an error because the adsorbent in Adsorbent-catalyst E is  $\beta$ -zeolite (95) which is not an adsorbent within the scope of the present invention.

Claims 7 and 26 have been amended to recite that the adsorbent of the adsorbent-catalyst used in the system of the present invention for purification of exhaust gas from an internal combustion engine contains, as a main adsorbent component, an H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more. The amendment to recite that the H/ $\beta$ -zeolite is a main adsorbent component is supported in the specification on page 7, lines 9 to 13. The amendment to limit the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio to 110 or more is supported, inter alia, by the description of Adsorbent A in the examples which is an H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110.

In the Final Office Action dated May 31, 2005, the Office has maintained all of the rejections that were made in the previous Action (dated November 16, 2004) and, additionally, is rejecting

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claims 7-8, 11-12, 15, 17, and 29-30 under 35 U.S.C. 102(b) as being anticipated by JP-A-7-213910. JP-A-7-213910 was cited in the Information Disclosure Statement filed May 5, 2005.

Prior to responding to the rejections in the Action, applicants will explain below the gist of the present invention.

The present invention is directed to the use of  $\beta$ -zeolite (identified as H/ $\beta$ -zeolite in the claims of the present application), which has excellent adsorption capability, in in-line exhaust purification systems as disclosed in EP-A-602963, EP-A-661098 and the like, which are owned by the assignee of the present application.  $\beta$ -zeolite useful as an adsorbent in the present invention is defined by its  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio, taking thermal stability of the adsorbent into consideration. That is,  $\beta$ -zeolite having a specified  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio as recited in the claims now on file in the present application, has a thermal stability which can retain the adsorption capability even when exposed to an exhaust gas of automobiles having a temperature of 750 to 850 °C for 100 hours under actual driving conditions.

The term "in-line" as used in the description and claims of the present invention means an arrangement manner of an exhaust purification system in an automobile wherein a catalyst, a catalyst-adsorbent, and a catalyst are aligned in series in the

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exhaust pipe of the automobile. The system aligned in this manner is different from one wherein an adsorbent is provided in a bypass pipe and the system is operated by switching a valve to cause the HC contained at the cold start to be adsorbed by an adsorbent provided in the bypass pipe. This latter type of system is disclosed in JP-A-2-2-75327, SAE Paper No. 920847, and the like.

The in-line system of the present invention is disclosed in EP-A-602963, EP-A-661098, and JP-A-7-213910, cited in the rejections in the present application. WO 94/11623 also describes an in-line type of exhaust system although preferred embodiments are directed to a system requiring a heat exchange means.

Prior Art vs. Present invention

(1) EP-A-602963 and EP-A-661098

These documents disclose the use of zeolites having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 20 or more (Si/Al ratio of 40 or more) in the purification systems disclosed therein. Various types of zeolites inclusive of ZSM-5, USY,  $\beta$ -zeolite, silicalite, and metallosilicate are exemplified. However, there are no concrete examples wherein  $\beta$ -zeolite is used as an adsorbent therein. Moreover, these documents are silent concerning the thermal durability of any of the zeolites disclosed therein.

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These documents also disclose the combination of a noble metal as a catalyst and an adsorbent wherein a zeolite other than  $\beta$ -zeolite is used as the adsorbent, i.e., a catalyst-adsorbent. However, again there are no concrete data as to the thermal durability of the zeolites used therein.

As can be understood from the above discussion, there is in EP-A-602963 and EP-A-661098 nothing to suggest the importance of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in zeolites to judge thermal durability as an index for long-lasting adsorption capability after exposure for a long period of time to an actual exhaust gas.

(2) JP-A-7-213910

This document discloses a catalyst-adsorbent wherein various zeolites are suggested for use as the adsorbent. In the working examples, a catalyst-adsorbent comprising a noble metal as a catalyst and  $\beta$ -zeolite having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 100 as an adsorbent is described. The significance of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of the zeolites is described in paragraph [0012] of the document as follows:

"There are various kinds of zeolites, in the present invention, it is preferable to choose, as a zeolite usable for the present inventive method, those having a sufficient HC adsorption ability and high durability in a wide range of temperature from normal (ambient) temperature to a relatively high temperature in the

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atmosphere wherein water is present. Among them, it is preferable to use a zeolite having  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 50 to 2000. For example, mordenite, USY,  $\beta$ -zeolite, and ZSM-5 can be illustrated. If the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of mordenite,  $\beta$ -zeolite, ZSM-5 and USY is less than 50, the zeolite can not adsorb effectively hydrocarbons contained in the exhaust gas because water co-presented therein hinders the adsorption of hydrocarbons. On the other hand, if the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of mordenite,  $\beta$ -zeolite and ZSM-5 exceeds 2000, or USY exceeds 300, respectively, the hydrocarbon adsorption ability of the zeolite decreases. It is more preferable to mix two or more of zeolites having different pore sizes or pore structures to adsorb effectively various kinds of HCs in the exhaust gas."

As can be understood from the above description, the criticalness of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of the zeolites in the invention disclosed in JP-A-7-213910 is described only in terms of hindrance of hydrocarbon adsorption by water molecules in the exhaust gas and decrease in hydrocarbon adsorption. That is, JP-A-7-213910 discloses nothing concerning the significance of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio with respect to thermal durability of the zeolites when exposed to high temperature for a long period of time. In fact, there are no data as to the thermal durability of any of zeolites used therein.

(3) WO 94/11623

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This document teaches the use of  $\beta$ -zeolite having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 25 to 500 in an apparatus for treating an engine exhaust gas. However, it does not mention the use of the zeolite as a catalyst-adsorbent. (The present application is restricted to a catalyst-adsorbent where a  $\beta$ -zeolite having a specified  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio is used as the adsorbent). The  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio in WO 94/11623 is used merely to define the acidity. Indeed, there are no data regarding thermal durability described in WO 94/11623.

It is further noted that the working examples in WO 94/11623 are directed to a system provided with heat exchange means. There are no working examples wherein the system is not provided with heat exchange means.

Criticalness of  $\text{SiO}_2/\text{Al}_2\text{O}_3$  Ratio in the Present Invention

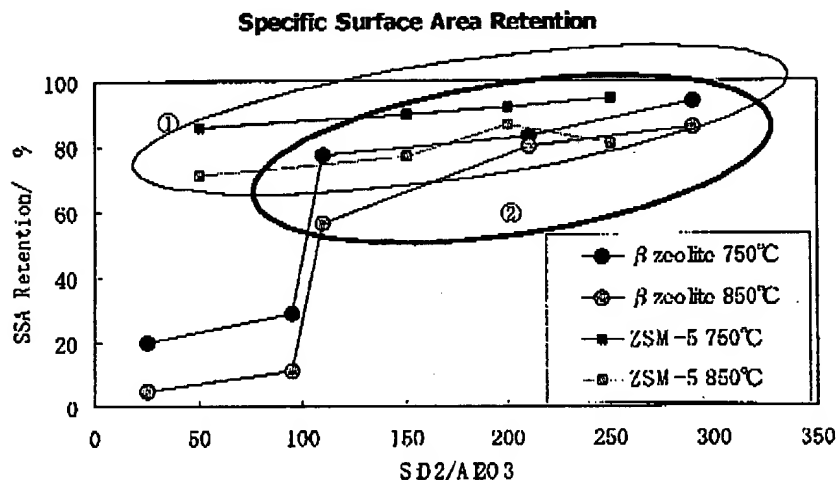
The present invention is directed to an exhaust gas purification system using  $\beta$ -zeolite having excellent adsorption capability for use in an adsorbent-catalyst in the system. As a result of extensive studies, the applicants found that  $\beta$ -zeolite having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 100 or more and, especially, 110 or more as is now claimed, can show unexpectedly good persistence in the adsorption capability of HC even when exposed to a high temperature for a long period of time in an in-line system, as is demonstrated by the data in Table 1 of the present application.

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None of the references cited by the Office teaches or suggests criticalness of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio with respect to thermal durability.

The criticalness of the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of the  $\beta$ -zeolite with respect to thermal durability is illustrated in the following chart.



As can be taken from the chart, it has been confirmed that a  $\beta$ -zeolite, the  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of which is 110 or more and within the range circled by circle 2, can exhibit unexpectedly superior thermal durability in the system for exhaust gas purification of the present invention. ZSM-5, having a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio within the range circled by circle 2, exhibits good thermal stability but has inferior adsorption capability.

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It is noted that the above chart is based on the data of Table 1 of the declaration under 37 C.F.R. § 1.132 submitted with the response filed in this application on April 21, 2003, a copy of which is enclosed herewith for the convenience of the Office. The data of Table 1 of the declaration under 37 C.F.R. § 1.132 filed on April 21, 2003, were calculated from the thermal durability data identified in Table 1, page 21, of the present application. The thermal durability data of Table 1 of the application were obtained using the method described on page 13, lines 4 to 15, of the application.

Rejections

Referring to the rejections of the claims in the Action of May 31, 2005, and, first, to the rejection of the claims as being anticipated under 35 U.S.C. § 102(b) by the disclosure of JP 07-A-213910, this rejection has been overcome by the amendments to claims 7 and 26 to recite that the adsorbent of the adsorbent-catalyst of the in-line exhaust gas purification system of the present invention contains an H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more.

Referring, second, to the rejections of the system for purification of exhaust gas from an internal combustion engine of the claims of the present application as being obvious under 35



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U.S.C. § 103(a) over the combinations of WO 94/11623 in view of EP-A-661098, EP-A-602963 and JP-A-7-124428; EP-A-661098 in view of WO 94/11623; EP-A-602963 in view of WO 94/11623; EP-A-602963 in view of WO 94/11623 in further view of EP-A-661098 and JP-A-7-213910 in view of EP-A-661098, applicants respectfully submit that none of these combinations provides the necessary motive to a person of ordinary skill in the art to pick and choose H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more for use as an adsorbent of an adsorbent-catalyst in an in-line exhaust system.

In WO 94/11623,  $\beta$ -zeolite is used merely as an adsorbent. In the case of an adsorbent, the  $\beta$ -zeolite is not exposed to a higher temperature, compared with the case wherein  $\beta$ -zeolite is used together with a catalyst like the one disclosed in EP-A-602963, for example.  $\beta$ -Zeolite is exposed to a higher temperature when used together with a catalyst because the combustion of HC adsorbed during a cold start repeatedly occurs. Thus, since there is no description as to thermal durability in WO 94/11623, an ordinary artisan would not be motivated to use  $\beta$ -zeolite disclosed in WO 94/11623 in an in-line system as disclosed, for example, in EP-A-602963. In this respect, it is noted that the heat exchange means which definitely moderates the heat generated by combustion of the HC adsorbed is employed in all of the working examples of WO

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94/11623. Therefore, without having a proper knowledge as to the thermal durability of  $\beta$ -zeolite, an ordinary artisan could not reasonably predict the effects of the use of H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more as an adsorbent of an adsorbent-catalyst in an in-line exhaust system.

The references cited by the Office, therefore, alone of in any combination, cannot properly support a case of prima facie obviousness under 35 U.S.C. § 103(a) of the claims of the present application.

Notwithstanding the insufficiencies of the references to support prima facie obviousness of the in-line exhaust gas purification system of the present invention which includes an adsorbent-catalyst in which the adsorbent contains an H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more, the data of Table 1 of the present specification and that of the 132 declaration filed April 21, 2005, show that the H/ $\beta$ -zeolite having an  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 110 or more provides unexpectedly superior thermal durability such that the adsorbent can maintain high adsorption capability of HC even when exposed to high temperatures for a long period of

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time. These data are sufficient to rebut any prima facie obviousness alleged to be supported by the prior art.

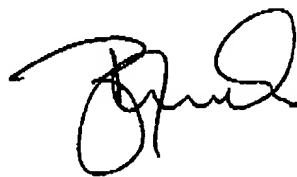
Removal of the 35 U.S.C. § 102 and 35 U.S.C. § 103(a) rejections of the claims is believed to be in order and is respectfully requested. Issuance of a Notice of Allowance is also requested.

In the event that this paper is not considered to be timely filed, applicants hereby petition for an appropriate extension of time. The fee for any such extension may be charged to our Deposit Account No. 111833.

In the event any additional fees are required, please also charge our Deposit Account No. 111833.

Respectfully submitted,

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Table 2

	Position of system mounting* (mm)	Order of mounting of system components**	Hydrocarbons reduction from 0 to 150 seconds (%)	Total hydrocarbons emission in FTP (g/mile)
sample 1	600	Catalyst A→Adsorbent-catalyst C→Catalyst B (850°C) (850°C) (850°C)	78	0.046
sample 2	600	Adsorbent-catalyst A→ Catalyst C (850°C) (850°C)	72	0.059
sample 3	1000	Catalyst A→Adsorbent E→ Catalyst B→ Catalyst C (850°C) (750°C) (750°C) (750°C)	65	0.055
sample 4	1000	Adsorbent-catalyst B→ Catalyst C (750°C) (750°C)	62	0.059
sample 5	1000	Catalyst C→Adsorbent D→ Catalyst A→ Catalyst B (850°C) (850°C) (850°C) (850°C)	66	0.049
sample 6	1000	Adsorbent B→ Catalyst C (750°C) (750°C)	63	0.072
sample 7	1000	Catalyst D→ Adsorbent-catalyst [(E)] G→ Catalyst D (850°C) (850°C) (850°C)	80	0.042
sample 8	800	Catalyst D→ Adsorbent-catalyst H→ Catalyst D (850°C) (850°C) (850°C)	82	0.039
sample 9	800	Catalyst D→ Adsorbent-catalyst I→ Catalyst D (850°C) (850°C) (850°C)	81	0.040
comparative sample 1	600	Catalyst A→Adsorbent-catalyst G→Catalyst B (850°C) (850°C) (850°C)	35	0.108
comparative sample 2	600	Catalyst A→Adsorbent O→ Catalyst B (850°C) (850°C) (850°C)	39	0.098
comparative sample 3	600	Catalyst A→ Adsorbent-catalyst F→ Catalyst B (850°C) (850°C) (850°C)	46	0.090

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mparative ample 4	1000	Catalyst A→Adsorbent-catalyst G→Catalyst B→Catalyst C (850°C) (750°C) (750°C)	40	0.108
mparative ample 5	1000	Catalyst A→ Adsorbent-catalyst E→ Catalyst B→ Catalyst C (850°C) (750°C) (750°C)	28	0.137
mparative ample 6	1000	Adsorbent J→ Catalyst C (750°C)	12	0.186
mparative ample 7	1000	Adsorbent H→ Catalyst C (750°C)	28	0.104

distance from engine exhaust port to point of system closest to said port.

Order starting from the most upstream component in the direction of exhaust gas flow. Figures in parentheses indicate an inlet temperature during durability test.